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Translation of Priority Document

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METHOD FOR FABRICATING HIGH-PURITY SILICA GLASS USING SOL-GEL PROCESSING

BACKGROUND OF THE INVENTION

1. Field of the Invention

The present invention relates generally to a fabrication method of silica glass, and in particular, to a method for fabricating high-purity silica glass using a sol-gel processing.

2. Description of the Related Art

Silica glass, which is a raw material of high-purity glass articles such as photomask and optical fiber as used in fabrication of semiconductor devices, is generally manufactured using a natural quartz processing, a synthetic quartz processing or a sol-gel processing.

The sol-gel processing, inter alia, is a technique of securing high purity of the starting material to provide high purity and uniformity of the molecule units and involving a liquid phase method that allows high productivity and relatively arbitrary control of the composition and structure of the products compared to the other processings. Such a sol-gel processing is also economical because almost all steps but sintering are carried out at a low temperature.

A fabrication method of silica glass using the sol-gel processing is disclosed in detail in U.S. Patent No. 5,240,488 under the title of "Manufacture of vitreous silica product via a sol-gel process using a polymer additive".

In the fabrication of silica glass using the sol-gel processing, there are vari us factors including temperature, composition, pressure, acidity and solvent

that that reciprocally act on one another in the transition of sol to gel and have an influence on the strength of the gel. To overcome the problem, an attempt has been made on a method for securing the flexibility and strength of the gel with prevention of cracks in the drying step.

As disclosed in U.S. Patent No. 5,240,488, the fabrication method of silica glass using the sol-gel processing involves dispersing a fumed silica in an alkaline region to form a sol, adding a polymer and effecting a gelation of the sol to produce the final silica glass. This method may prevent creation of cracks in the gel during the drying step and reduce the residual carbon of the final product caused by addition of an organic binder, thus allowing production of large-sized silica glass. However, there is still a risk of crashing the gel in the manufacturing process because the gel has a low strength.

Meanwhile, the sol-gel processing involves addition of a fluorine compound so as to reduce the refractivity and heat expansion coefficient of silica as well as the viscosity of the silica glass at a high temperature. The fluorine compound is an acidic substance used as a gelation agent for a polymeric sol or a silica particulate sol, which are both dispersed in an acidic region with a low hydrogen ion content. The reason why the existing fluorine compounds cannot be used as a gelation agent in the sol dispersed in the alkaline region is that adding the fluorine compounds to the sol dispersed in the alkaline region may lower the hydrogen ion content of the sol and incur a rapid gelation.

SUMMARY OF THE INVENTION

It is, therefore, an object of the present invention to provide a method for fabricating high-purity silica glass using a sol-gel processing in which a fluorine compound is added to provide bar-shaped silica glass with minimized cracks during the drying step and improved plasticity.

It is another object of the present invention to provide a method for fabricating high-purity silica glass using a sol-gel processing that allows production of a particulate sol containing a fluorine compound even in the alkaline region so as to reinforce the wet gel, thereby minimizing formation of cracks in the drying step.

To achieve the above and other objects, there is provided a method for fabricating high-purity silica glass using a sol-gel processing. The method comprises the steps of: (a) mixing a deionized water with a fluorine compound and a dispersion agent to prepare an aqueous premix solution; (b) mixing the aqueous premix solution with a fumed silica; (c) mixing the resulting mixture to form a dispersed sol; (d) aging the dispersed sol at the ambient temperature to stabilize silica particles; and (e) removing air voids from the sol and adding a gelation agent.

BRIEF DESCRIPTION OF THE DRAWINGS

The above and other objects, features and advantages of the present invention will become more apparent from the following detailed description when taken in conjunction with the accompanying drawings in which:

FIG. 1 is a flow chart showing a process for fabricating high-purity silica glass using a sol-gel processing in accordance with the present invention.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENT

FIG. 1 is a flow chart showing a process for fabricating high-purity silica glass using a sol-gel processing in accordance with the present invention. As shown in FIG. 1, the fabrication method of high-purity silica glass using a sol-gel processing according to the present invention comprises the steps of aqueous

premix s luti n fabrication 100, mixing 200, dispersion 300, aging 400, defoamation 500, molding 600, gel aging 700, demolding 800, drying 900, low heat treatment 1000, and sintering 1100.

Unlike the conventional method where a polymer is added as a binder to an aqueous premix solution of silica and deionized water, the present invention method for fabricating high-purity silica glass using a sol-gel processing uses a fluorine compound to prepare an aqueous premix solution, mixes the aqueous premix solution with a fumed silica and subjects the mixture to dispersion under the alkaline condition and ageing to form a sol, which is then mixed with a gelation agent to change the sol to the gel.

The aqueous premix solution fabrication step 100 is mixing a deionized water 102 with a fluorine compound 104 and a dispersion agent 106 to prepare an aqueous premix solution. This step is to prevent a rapid gelation of the sol that may otherwise happen due to direct addition of the fluorine compound 104. Examples of the fluorine compound as used herein include NH_4F, $(NH_4)SiF_6$ and HF. The dispersion agent 106 is tetramethylammonium hydroxide that maintains the hydrogen ion content of the aqueous premix solution in the range of 10 to 13, with enhanced dispersability of silica particles. The fluorine content of the silica glass can be readily controlled by regulating the amount of the fluorine compound 104 added in the aqueous premix solution fabrication step 100.

The mixing step 200 is mixing the aqueous premix solution with a fumed silica 202. The amount of the fumed silica added is in the range of 40 to 60 wt.%.

The dispersion step 300 is mixing the mixture obtained in the mixing step 200 to form a dispersed sol. This step can be carried out in a high shear mixer and followed by ultrasonic milling and centrifugal separati n for the purpose of increasing the uniformity of the silica particles and removing

impurities.

The aging step 400 is aging the sol of the dispersion step 300 at the ambient temperature for a predetermined time to stabilize the silica particles in the sol.

The defoamation step 500 is removing the air voids contained in the aged sol and adding a gelation agent 502. The gelation agent 502 controls the acidity of the sol and incurs gelation of the sol, thus reducing the gelation time with increased strength of the wet gel. Examples of the gelation agent 502 include methyl formate, methyl lactate and ethyl lactate, which are used alone or in combination.

The molding step 600 is pouring the sol of the defoamation step 500 into a mold of a defined shape and subjecting it to polymerization reaction and gelation. The mold has a cylindrical shape for an article molded in the rod form like a core rod and additionally includes a core rod to provide a molded article in the form of a tube including an overjacketing tube or a substrate tube.

The gel aging step 700 is aging the molded article in the mold so as to increase the strength of the wet gel.

The demolding step 800 separates the wet gel from the mold. In some cases, this step 800 can be carried out using the water pressure in a water bath in order to prevent damage of the wet gel.

The drying step 900 is drying the wet gel separated from the mold in the demolding step 800 in a constant temperature and humidity chamber to form a dry gel.

The low heat treatment step 1000 is heat-treating the dry gel under th atmosphere of a gas such as chlorine, hydrogen or oxygen to decompose the residual water and organic substances in the dry gel and remove metallic impurities and hydroxyl groups (OH). This step removes the impurities from the dry gel as described above and is also called "purification step".

The sintering step 1100 sinters the dry gel of the low heat treatment 1000 into vitreous gel at a high temperature and produces the final high-purity silica glass. The sintering 1100 is carried out by heating the dry gel up to 1400 °C with a furnace moving up and down in the sintering furnace under the helium (He) atmosphere and produces the final high-purity bar-shaped silica glass. The silica glass as obtained in the present invention contains a fluorine compound that lowers the viscosity of the silica glass to reduce the time and cost required to a high heat treatment combining step such as overjacketing.

Example 1

2.825 g of deionized water was mixed with 375 cc of an aqueous solution containing 25 wt.% of tetramethylammonium hydroxide and NH_4F. 3000 g of a fumed silica (Aerosil-OX50, Degussa) was added to the mixture, which was then dispersed with a high shear mixer to form a sol.

Subsequently, the sol separated from the mixer was aged at 18 °C for 10 hours. 5000 g of the aged sol was weighted and removed of air voids using a vacuum pump. 95 cc of ethyl lactate was then added to the defoamed sol.

After a second defoamation, the sol was poured into a mold to provide a molded wet gel. This wet gel was separated from the mold and dried in a constant temperature and humidity chamber at 30 °C and 75% relative humidity to obtain a dry gel. The dry gel was then subjected to a heat treatment in the

temperature range of 300 to 600 °C at a heating rate of 50 °C/hr for 3 h urs in order t eliminate residual water and additives from the dry gel.

The dry gel after the heat treatment was placed in a sintering furnace and kept at a raised temperature of 900 °C at a heating rate of 100 °C/hr for 2 hours. As such, the chlorine gas was supplied into the sintering furnace to remove the residual hydroxyl group (OH) from the dry gel. Subsequently, the sintering furnace was heated to 1400 °C at a heating rate of 100 °C/hr under the helium (He) atmosphere for one hour to obain high-purity silica glass.

Example 2

30 g of glycerin was added as a plasticizer to the sol of Example 1. The subsequent procedures were performed in the same manner as described in Example 1.

Example 3

2.825 g of deionized water was mixed with 375 cc of an aqueous solution containing 25 wt.% of tetramethylammonium hydroxide and $(NH_4)SiF_6$. 3000 g of a fumed silica (Aerosil-OX50, Degussa) was added to the mixture, which was then dispersed with a high shear mixer to form a sol.

The sol separated from the mixer was aged at 18 °C for 10 hours. 5000 g of the aged sol was weighted and removed of air voids using a vacuum pump. 95 cc of ethyl lactate was then added to the defoamed sol. The subsequent procedures were performed in the same manner as described in Example 1.

Example 4

30 g of glycerin was added to the sol of Example 3. The subsequent procedures were performed in the same manner as described in Example 3.

The method for fabricating high-purity silica glass using a sol-gel processing in accordance with the embodiment of the present invention enables production of high-purity silica glass from a low-purity fumed silica and secures high strength of the wet gel, thus allowing production of large-sized silica glass.

Furthermore, the present invention method involves addition of the fluorine compound to the aqueous premix solution instead of adding the fluorine compound directly to the sol, so that the sol can secure dispersability with the prevention of rapid gelation.

While the invention has been shown and described with reference to a certain preferred embodiment thereof, it will be understood by those skilled in the art that various changes in form and details may be made therein without departing from the spirit and scope of the invention as defined by the appended claims.

WHAT IS CLAIMED IS:

1. A method for fabricating high-purity silica glass using a sol-gel processing, comprising the steps of:

(a) mixing a deionized water with a fluorine compound and a dispersion agent to prepare an aqueous premix solution;

(b) mixing the aqueous premix solution with a fumed silica;

(c) mixing the resulting mixture to form a dispersed sol;

(d) aging the dispersed sol at the ambient temperature to stabilize silica particles; and

(e) removing air voids from the sol and adding a gelation agent.

2. The method as claimed in claim 1, wherein the fluorine compound added in the preparation step (a) of the aqueous premix solution is NH_4F.

3. The method as claimed in claim 1, wherein the fluorine compound added in the preparation step (a) of the aqueous premix solution is $(NH_4)SiF_6$.

4. The method as claimed in claim 1, wherein the fluorine compound added in the preparation step (a) of the aqueous premix solution is HF.

5. The method as claimed in claim 1, wherein the dispersion agent used in the mixing step (b) is tetramethylammonium hydroxide.

6. The method as claimed in claim 1, wherein the gelation agent used in the defoaming step (e) is any one selected from the group consisting of methyl formate, methyl lactate and ethyl lactate.

ABSTRACT OF THE DISCLOSURE

Disclosed is a method for fabricating high-purity silica glass using a sol-gel processing that includes the steps of: (a) mixing a deionized water with a fluorine compound and a dispersion agent to prepare an aqueous premix solution; (b) mixing the aqueous premix solution with a fumed silica; (c) mixing the resulting mixture to form a dispersed sol; (d) aging the sol at the ambient temperature to stabilize silica particles; and (e) removing air voids from the sol and adding a gelation agent.